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INVESTIGATION OF COLD CATHODES FOR LONG LIFE CO2 WAVEGUIDE LASE--ETC(U)
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Electrical Engineering Department

UNIVERSITY OF MARYLAND, COLLEGE PARK, MD 20742

Investigation of Cold Cathodes

for

Long Life CO_2 Waveguide Lasers

Progress Report and Proposal
for Contract N00014-79-C0312

Submitted to:

Dr. Nicolay

ONR

by

U. Hochuli

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COLLEGE OF ENGINEERING: GLENN L. MARTIN INSTITUTE OF TECHNOLOGY

38441

INTRODUCTION

We have proposed to solve the life problems of the low power longitudinally DC excited, CW wave guide laser. The problem areas that presently limit the life of this laser are listed below:

1. The requirement of a cold cathode technology that allows one to maintain the proper gas composition over a long period of time. These cathodes have to have a low sputtering rate and produce deposits that are stationary and well attached to harmless areas in the neighborhood of the cathode. No mirror deposits can be tolerated.

2. Mirror intensity limits have to be known.

3. Proper sealing techniques are absolutely necessary for long actual laser-life as well as shelf life.

A detailed discussion of our present research results in these areas follows.

1. Cold Cathode Technology

In our proposal we predicted that the ten times higher pressure of the waveguide laser would increase the $10 mA/cm^2$ cathode current density of the low pressure laser to roughly $1 A/cm^2$. Actual cathode current densities, calculated by observing the cathode spot area on a flat NiO cathode are shown in figure 1.

These results indicate about $330 mA/cm^2$ for a gas pressure of 200 Torr and a flat cathode. In a hollow cathode we may expect higher current densities. Attempts to lower this current density by heating the cathode surface were partially successful.

By observing the cathode spot on external cylinder cathodes with a hemispherical end closest to the anode we found the existence of a critical cathode temperature. Exceeding this temperature suddently increases the cathode spot, for a given constant cathode current, 5 to 10 times. This leads of course to the wanted 5 to 10 fold decrease in cathode current density. This critical temperature is 600 to 800^{o}C for Cu cathodes and 300 to 1000^{o}C for Ni cathodes. At these temperatures, the copper cathodes have an extremely low sputtering rate while the nickel cathodes sputter considerably more. In both cases the sputtering products attach themselves to the colder surfaces on the cathode itself or on the walls in the vicinity of the cathode. Gas analysis results for discharge tubes with these cathodes are shown in figures 2 to 5. Despite carful construction, the power generated by the cathode fall-voltage cathode-current product is not sufficient for self heating. An additional 3.5 to 13 watts had to be generated by insulated Pt wire heaters around the cathodes to bring them up to the critical temperatures. We have tried in vain to design cathodes small enough to self-heat. Cathodes with volumes of the order of 1 mm^3 do self-heat but have an insufficient area to accomodate the cathode spot with the lower current density. Hesitantly we have to admit that these indirectly heated cathodes do not offer a good, viable engineering solution. Instead, we have turned to semiconducting metal oxides for cold cathodes. Discharge tube results are so far quite encouraging. Figures 6 to 9 show reasonable gas composition results. The sputtering rates are relatively low and the sputtering products seem to be attached to surfaces near the cathode.

We had also proposed to investigate the use of copper hydroxide as a water vapor source:

$$Cu(OH)_2 \underset{T}{\leftrightarrow} CuO + H_2O.$$

The published H_2O vapor pressure is supposed to be around 1 Torr at 20°C.

We found copper hydroxide to be a highly unstable H_2O vapor source. After evacuation and sealing-off the H_2O vapor pressure indeed rose to about 1 Torr, but within 4 to 6 weeks we found that the pressure had risen so much that we also found liquid water on the walls and all the bluish looking Cu $(OH)_2$ had turned black, an indication of conversion to CuO. It is possible that this could be prevented by avoiding day light expoure or it would not happen in the presence of the laser gas. From an engineering point of view we consider $Cu(OH)_2$ too unstable to be used as a reliable H_2O vapor source.

2. Mirror Intensity Limits

Accurate mirror damage threshold information is very difficult to find or may nonexistent for particular experimental conditions. Mirrow manufacturers usually do not test their own mirrors and often depend on feedback from their customers. Cautious estimates run around $1KW/cm^2$ at 50°C blank temperature, if a laser life in excess of several thousand hours is required.

In order to stay within this limit a small waveguide laser with a peak output intensity of only $1W/mm^2$ has to be restricted to an output mirror with a reflectivity of $R<0.9$.

We have destroyed mirrors working with $2KW/cm^2$ in trying to tune the cavity length by heating the mirror supports to $30^{o}C$. These mirrors lasted less than 500h. Mirrors working at room temperature with intensities of $1KW/cm^2$ have so far shown no measurable damage over periods up to 3000 hours.

3. Sealing Techniques

We have always been convinced that it is irresponsible to use epoxy resin seals in experiments that already have too many variables and which take a long time to show conclusive results. The indium seals we developed years ago are a very attractive alternative because they allow one to join materials with different thermal expansion coefficients while adding only a few microns in film thickness to the seal length. These seals are vacuum tight and do not contaminate other parts of the laser because of the extremely low vapor pressure of liquid indium. Their only drawback is the relatively low melting point of indium which,after assembly of the structure, limits outgassing temperatures to at most 150^{o}. The fact that the laser bodies are assembled in a vacuum at temperatures between 250^{o} and $300^{o}C$ helps to pre-outgas the structures during the sealing process. Until recently we have not been able to prevent very small leaks in the indium seals of our BeO laser structures. The very good heat conductivity of BeO requires one to make ‿ve indium seals simultaneously at the same temperature, namely: two vertical end plates and the three horizontal electrode flanges. An additional difficulty arises from the fact that the grain size of the sintered BeO does not permit one to polish these surfaces to better than about a 2-4μ inch surface roughness. In the past these adverse conditions have always resulted in very small leaks that had to be sealed with an external application of epoxy resin. A major effort, requiring about 5 man months of time, has now permitted up to perfect these sealing techniques to such a degree that no leaks can be detected on the most sensitive range of a Veeco MS-12 He-leak detector with the whole laser surrounded by He contained in a plastic gag. All the laser flanges are made from type 416 stainless steel sealed directly to BeO. The ZnSe windows are sealed to type 416 stainless flanges and all flanges use indium "O" rings for the final assembly and alignment. This is the first time we have properly assembled laser structures and the parts are shown in the pictures furnished in the appendix.

Laser Results

A power output versus time curve for a BeO laser structure with internal mirrors is shown in figure 10. The laser structure used was similar to the one shown in the appendix but the seals were not yet perfected and had to be repaired with an external application of epoxy resin. The first run marked I, shows that the laser output drops from 1.2W to to about 1W during the first 500 hours, to 0.6 after 1000 hours and then slowly decreases to 0.32W after 1600 hours. Gas analysis with the mass spectrometer gave no clue for this drop. Vacion pump-down to about 10^{-6} torr over a period of a few hours and a new filling restores the original power but the laser output drops to 50% in 120 hours. Vacion pump-down to 10^{-8} torr over a period of 1 week with the laser heated to $80^{o}C$ are the basis for curve II in figure 10. As seen, the initial power output is restored to 1.2W and drops to 0.68W after 1680 hours. Again the mass spectrometer analysis of the gas gave no reason for this power drop. Pump-down to about 1 torr and a new gas fill yields 0.73W output power, barely a 10% power increase. This result substantiates the mass spectrometer analysis, indicating that the gas composition is not responsible for the power drop. This power drop could be explained by condensation products on the laser mirrors which introduce loses, but do not permanently damage the mirrors themselves and can again be removed by extensive evacuation. We have not been about to find any unusual outgassing products with our UTI mass spectrometer in the 0-300 AMU range. The cathodes of this laser were made from 4N pure copper and a quick search in the tables of the Handbook of Physics and Chemistry reveals the underligned compounds shown in the appendix that can be formed with Cu,C, O and H. Quite a few of these compounds are indeed relatively unstable and there may be others such as copper carbonyl that are so unstable that this particular source does not even list it and its existence is questioned in the literature. We are of course also aware of the claims that CO stored at high pressure in steel tanks can form such compounds as iron carbonyls and that filters are used for cleaning purposes. We have outgassed the laser again for one week to a residual vacion pump pressure of 10^{-8} torr and refilled the laser with the gas mixture. This time the gas mixture was slowly driven through a 10cm long 1cm ID tube filled with quartz wool heated to 400°C in an effort to remove such "gas bottle residues". Curve III in figure 10 shows that the filtering effort was futile. The resulting curve falls between curves I and II and can be best explained by the fact that the outgassing was not done as well as for run II because the structure was kept at room temperature instead of at 80°C as was done for run II. The mirror contamination hypothesis becomes even more convincing if we compare the previous results with the ones in figure 11. Here a similar laser structure which used Brewsters windows and external mirrors indicates an almost constant power output of 0.36W over a period of 2000 hours. These Brewster angle windows were 2 cm farther away from the laser bore ends than the internal mirrors which were separated by less than 0.5mm from these ends. The active discharge itself approaches the bore ends to within 1.6cm as shown by the laser body drawings in figure 12. The closer internal mirrors are of course much more prone to contamination or ion damage than the more distant Brewster windows. The argument above is weakened by the fact that a different type of cathode was used in this laser.

If the mirror contamination theory is correct we will have to investigate the following possible causes:

a) The creation of unsuitable cathode sputtering by products which end up on the mirrors.

b) The residual H or H_2O caused by poor outgassing which might allow the formation of these mirror deposits.

c) The epoxy coated seals which outgas or permit H_2O to penetrate into the laser.

d) The electrostatic charge accumulation on the mirrors and their insulated supports which attract the sputtering by-products.

e) The mirrors are possibly too close to the bore ends.

f) The laser, shown in figure 12, has built-in gas return paths that allow a slow gas circulation in front of the mirrors. This circulation may be responsible for carrying sputtering or decomposition products to the mirrors.

Conclusions

We have tested and partially evaluated over 40 combinations of cathode materials and gas fillings in discharge tubes. A few of these cathode materials are sufficiently promising to be tried in actual laser. We have also learned to assemble BeO laser structures with indium sealing techniques, assembled one structure and have two more ready for mounting. 20KW DC power supplies to feed six of these lasers have been build and two 30ℓ/s vacion pump systems were added to the existing one.

One laser, with epoxy coated seals, Cu cathodes and internal mirrors, produced 1.2W over the first 500h and dropped to half power after 1000 to 1500h as shown in figure 10. This power reduction seems to be the result of mirror deposits rather than gas mixture failure. Evacuation over prolonged periods of time restores the laser to the original condition. A similar laser structure, with Brewster windows and external mirrors shows in figure 11 an almost flat power output curve out to 2000 hours. This life improvement is most likely caused by the fact that the Brewster windows were farther away from the bore ends than the internal mirrors of the structure mentioned before.

Attempts will now be made to avoid this mirrow deposit problem by using laser structures with proper seals and other cathode materials.

<u>Proposal</u>

One of the most promising class of cathode materials consists of sintered and properly doped metal oxides. It may actually turn out that doping is more important to produce the proper cathode spot temperature than to directly affect the emissive properties of the cathode surface. The heat equation in the cathode material is given by

$$\mathrm{div}\ \lambda(\tau)\ \mathrm{grad}\ T = \frac{J^2}{\sigma(T)}$$

with the dominant boundary condition term for the cathode spot area:

$$\lambda(t) \operatorname{grad} T = V_c J,$$

and much more difficult boundary conditions for the rest of the cathode surface. An exact solution for the actual cathode geometry is probably quite hard to find but not essential to study the physics of the problem. Reducing the cathode to a cylinder of length ℓ, with a constant cross section equal to the cathode spot results in the one dimensional heat equation

$$\sigma(T) \frac{d}{dz} (\lambda(T) \frac{dT}{dz} = J^2 = \text{constant}$$

with the simplified boundary conditions

$$\lambda \frac{dT}{dz} = V_c J \text{ at } z = \ell$$

$$T = T_0 \text{ at } z = 0,$$

where

$\lambda(T)$ = T- dependent heat conductivity of the cathode material

$\sigma(T)$ = electrical conductivity of the cathode material

V_c = cathode fall voltage

J = cathode current density

It does not appear to be too difficult to solve the nonlinear differential equation numerically by an iterative method. The first step in this process, replacing $\lambda(T)$ and $\sigma(T)$ by constants, evaluated at an approximate average temperature, is good enough for the moment. This simplifies and linearlizes the equation to

$$\sigma\lambda \frac{d^2T}{dz^2} = J^2 = \text{const}$$

$$\lambda \frac{dT}{dz} = V_c J \quad \text{at } z = \ell$$

$$T = T_0 \quad \text{at } z = 0$$

Its solution

$$T(z) - T_0 = \frac{J^2 \ell}{\sigma\lambda} \left[\frac{z}{\ell} - \tfrac{1}{2} \left(\frac{z}{\ell}\right)^2 \right] + \frac{J V_c \ell}{\lambda} \quad \frac{z}{\ell}$$

yields

$$T(\ell) = \frac{J^2 \ell}{z\sigma\lambda} + \frac{J V_c \ell}{\lambda} + T_0$$

which can now be numerically evaluated. Typical values for the parameters are

$\lambda \approx 0.08$ J/cm°K

$\ell \approx 0.15$ cm

$J \approx 0.14$ A/cm^2

$V_c \approx 300$V

$\sigma \approx 10^{-2}$ to 10^{-7} $(\Omega\text{cm})^{-1}$ by doping.

With these values and with $T_0 \approx 200^\circ$C

$$T(\ell) \approx 185 \frac{10^{-4}}{\sigma} + 80 + 200 \ {}^\circ\text{C}$$

$$\approx 465^\circ\text{C for } \sigma = 10^{-4} (\Omega\text{cm})^{-1}.$$

The electrical conductivity can be expressed by

$$(T) = Ae^{-\frac{E}{kt}} .$$

This strong temperature sensitivity should have a highly stabilizing effect on the cathode temperature.

We feel this is certainly a far more promising way to reach the higher cathode temperature than indirect heating with a Pt wire heater.

Presently we propose to investigate this approach in discharge tubes and, if it appears to be successful, in actual lasers as well. At the same time we intend to continue testing some of the more promising cathodes in actual lasers. If the apparent mirror condensation problem persists with other cathode materials we may have to investigate and remedy its cause. In this case access to surface analysis equipment in government laboratories may have to be requested.

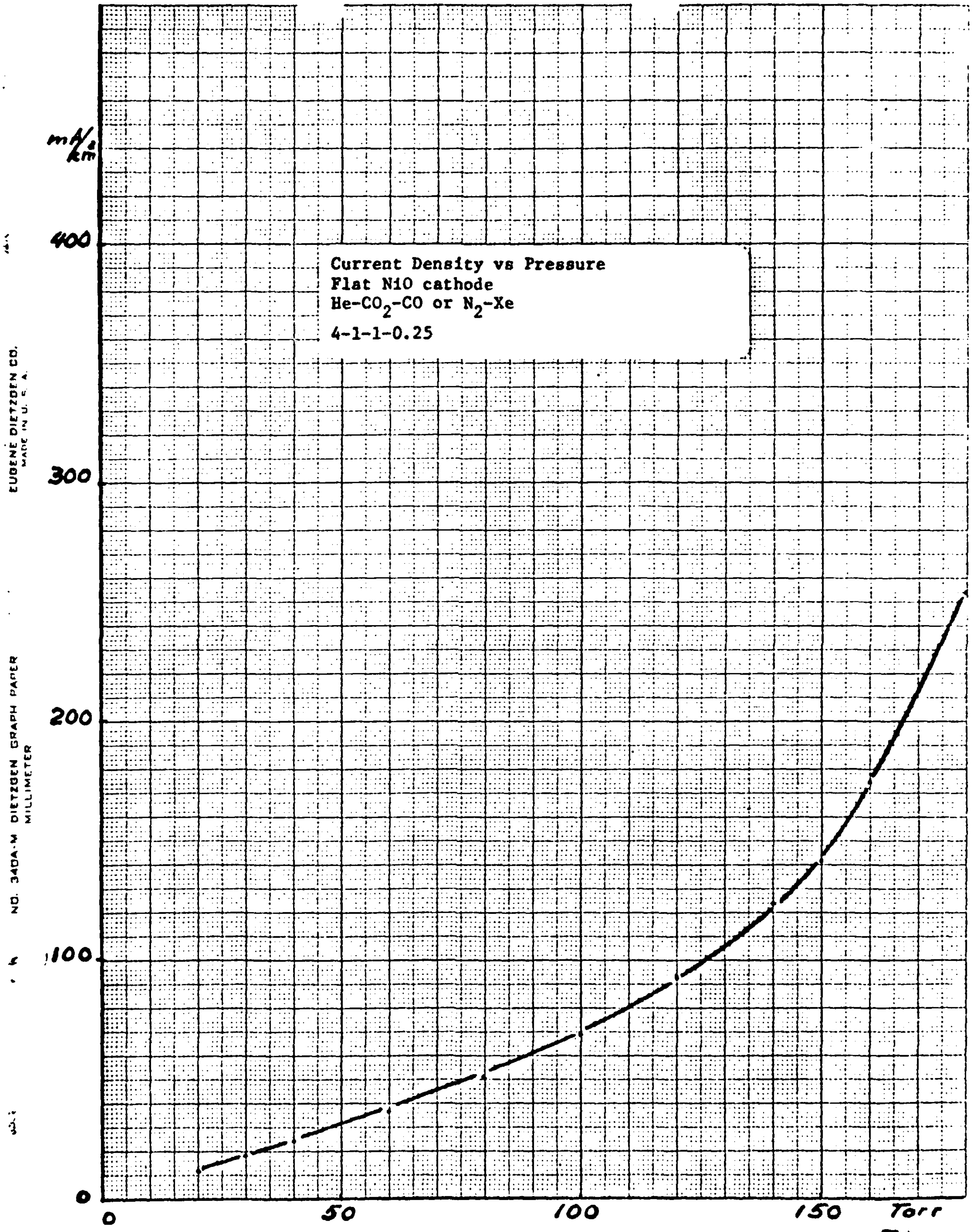

Current Density vs Pressure
Flat NiO cathode
He-CO_2-CO or N_2-Xe
4-1-1-0.25
mA/cm²
400
300
200
100
0
0
50
100
150
Torr
EUGENE DIETZGEN CO.
MADE IN U.S.A.
NO. 340A-M DIETZGEN GRAPH PAPER
MILLIMETER

Fig. 1

SEMI-LOGARITHMIC
3 CYCLES X 10 DIVISIONS PER INCH

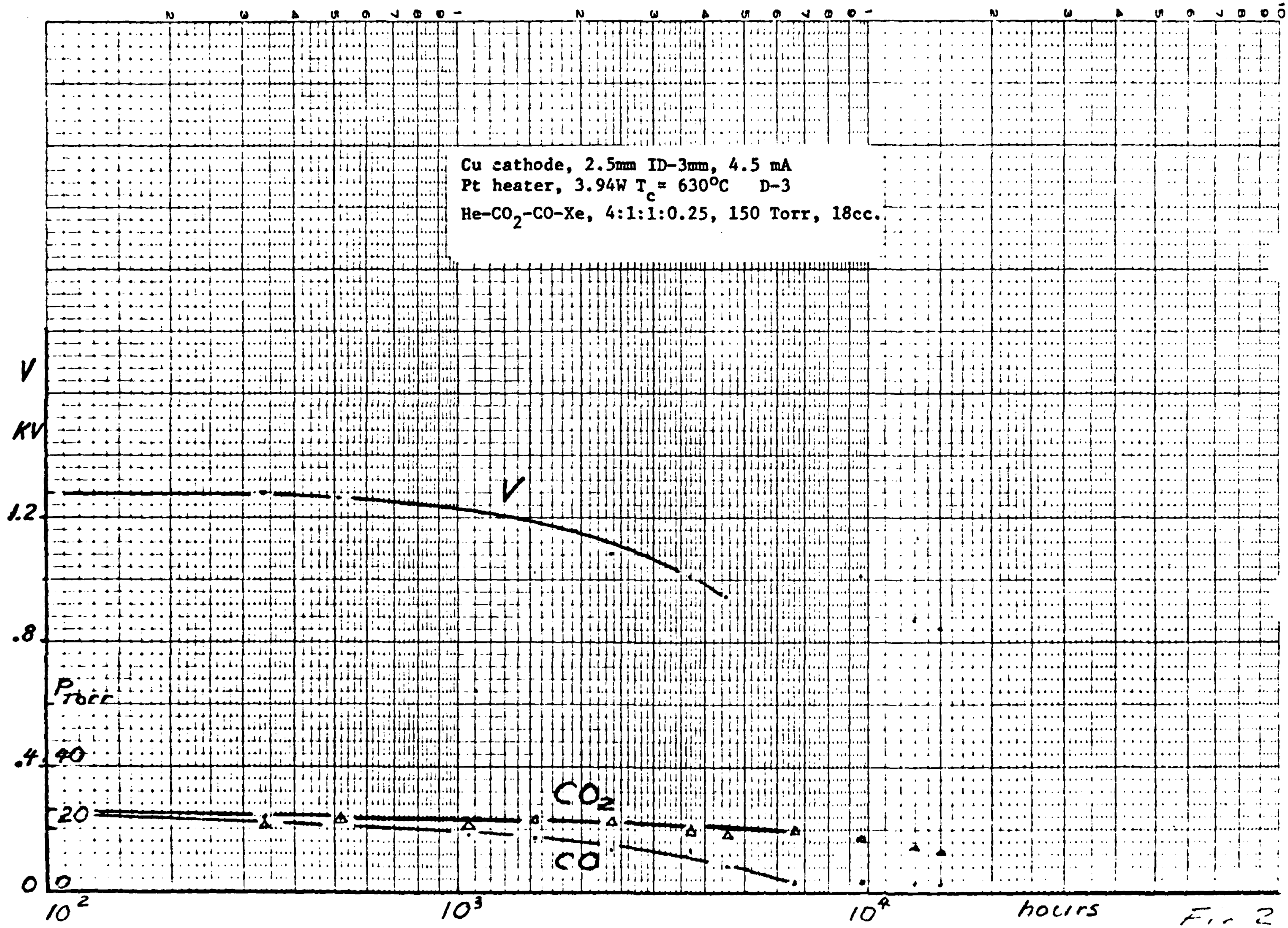


Fig 2

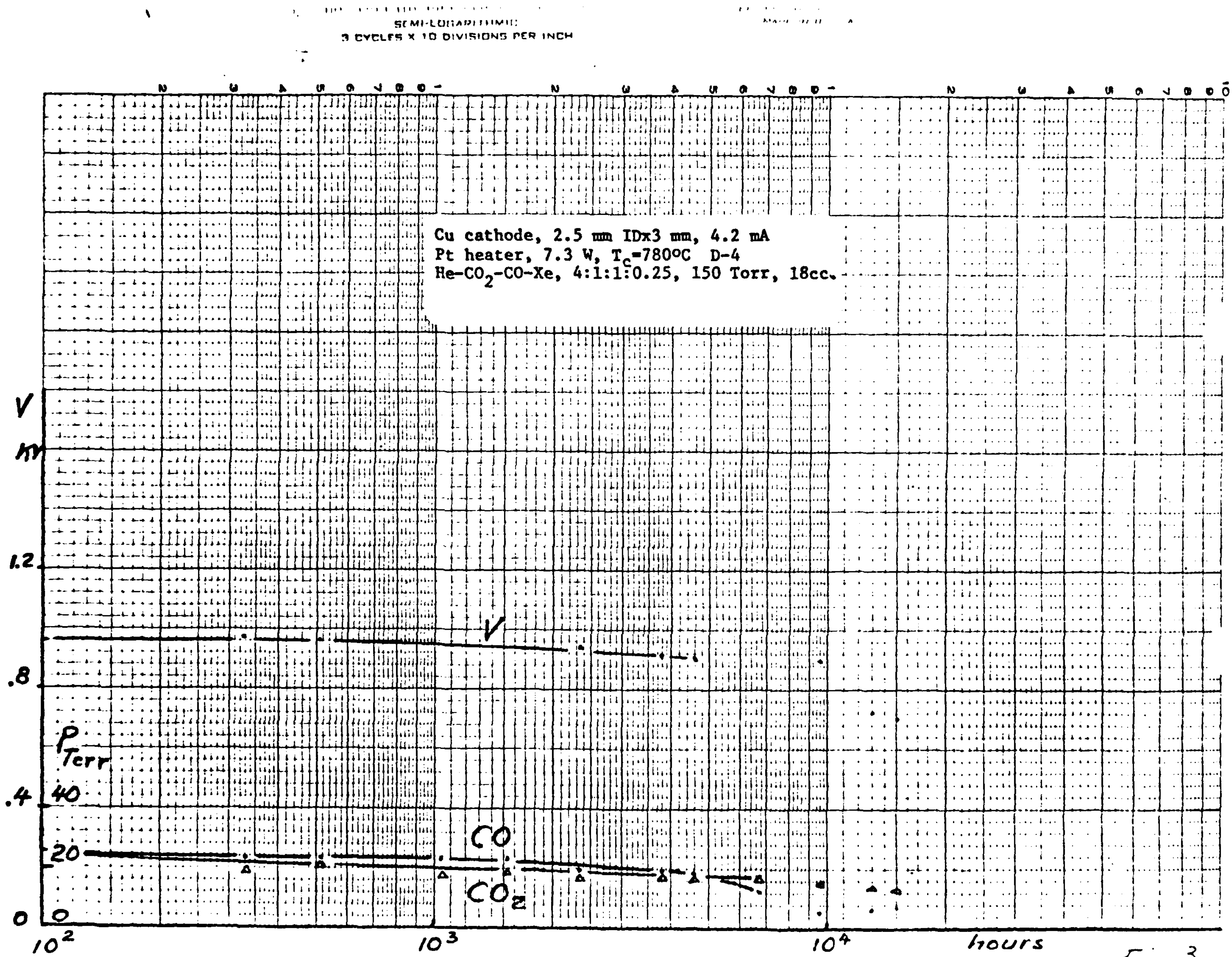

SEMI-LOGARITHMIC
3 CYCLES X 10 DIVISIONS PER INCH
Cu cathode, 2.5 mm IDx3 mm, 4.2 mA
Pt heater, 7.3 W, T_c=780°C D-4
He-CO_2-CO-Xe, 4:1:1:0.25, 150 Torr, 18cc.
V
kV
1.2
.8
.4
0
P
Torr
40
20
0
V
CO
CO_2
10^2
10^3
10^4
hours

Fig. 3

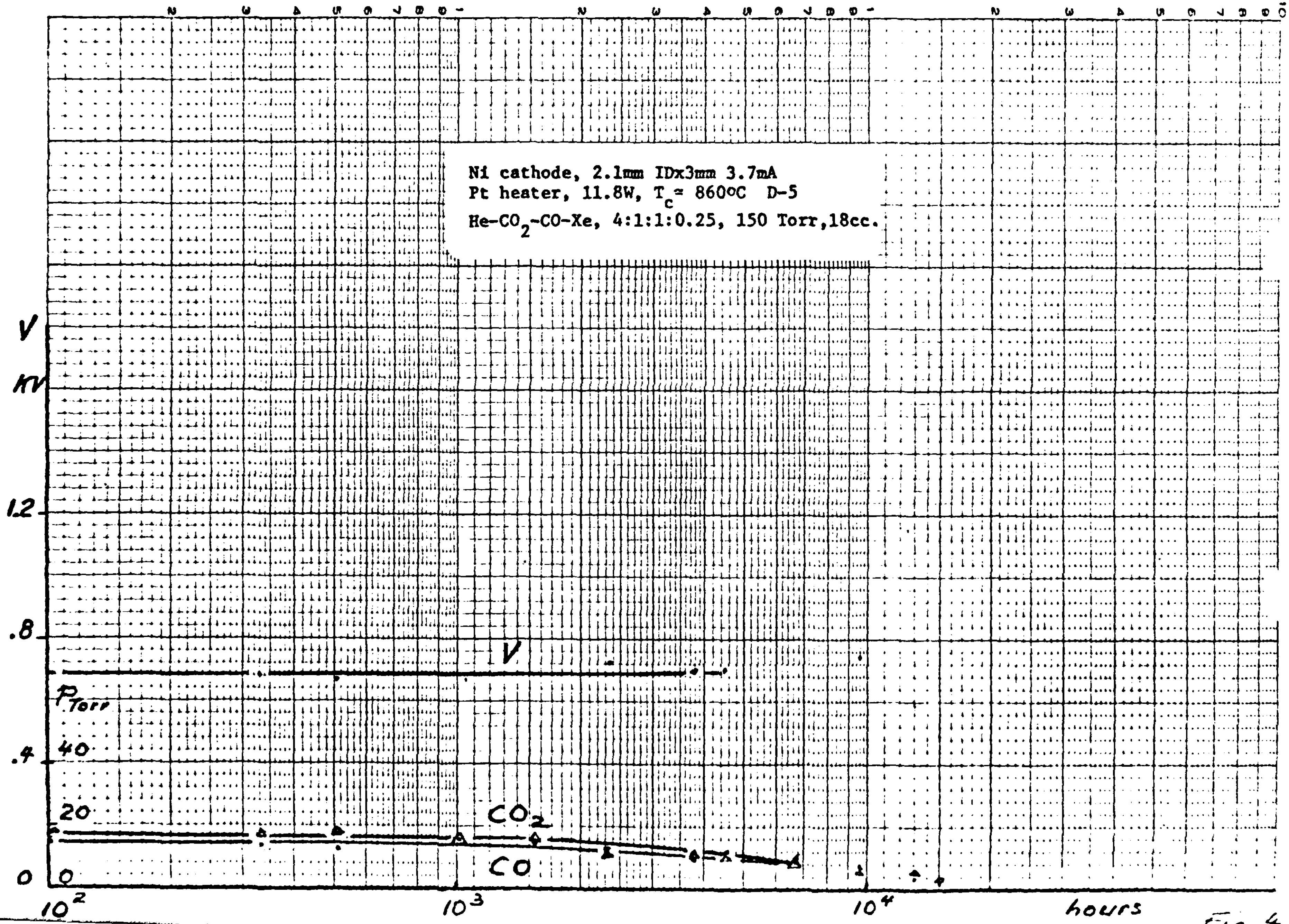

Ni cathode, 2.1mm IDx3mm 3.7mA
Pt heater, 11.8W, T_c = 860oC D-5
He-CO_2-CO-Xe, 4:1:1:0.25, 150 Torr,18cc.
V
KV
1.2
.8
.4
0
P_{Torr}
40
20
0
V
CO_2
CO
10^2
10^3
10^4
hours

Fig 4

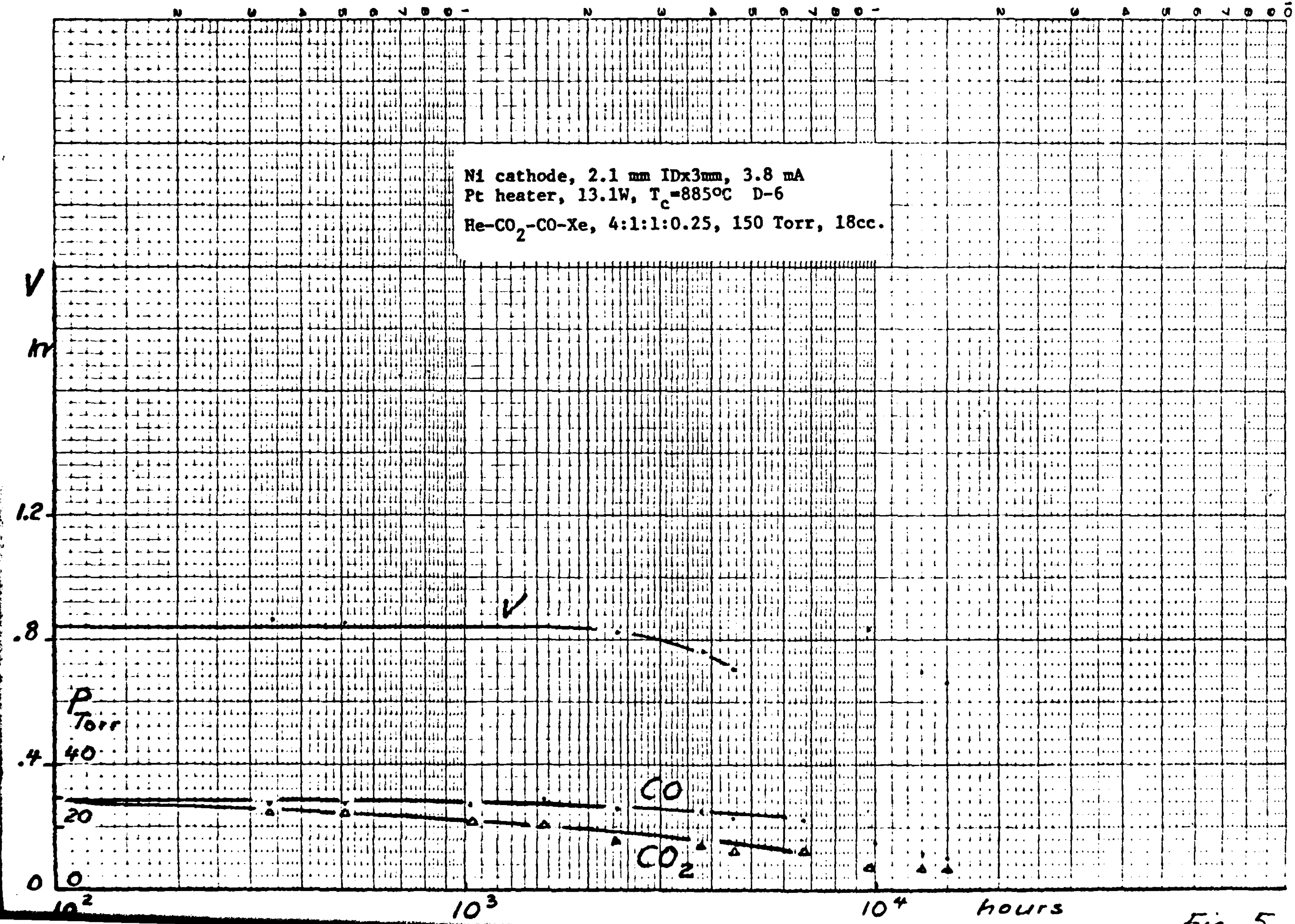

Ni cathode, 2.1 mm IDx3mm, 3.8 mA
Pt heater, 13.1W, T_c=885°C D-6
He-CO_2-CO-Xe, 4:1:1:0.25, 150 Torr, 18cc.
V
kv
1.2
.8
.4
0
P Torr
40
20
0
V
CO
CO_2
10^2
10^3
10^4
hours

Fig. 5

SEMI-LOGARITHMIC
3 CYCLES X 10 DIVISIONS PER INCH

MADE IN U. S. A.

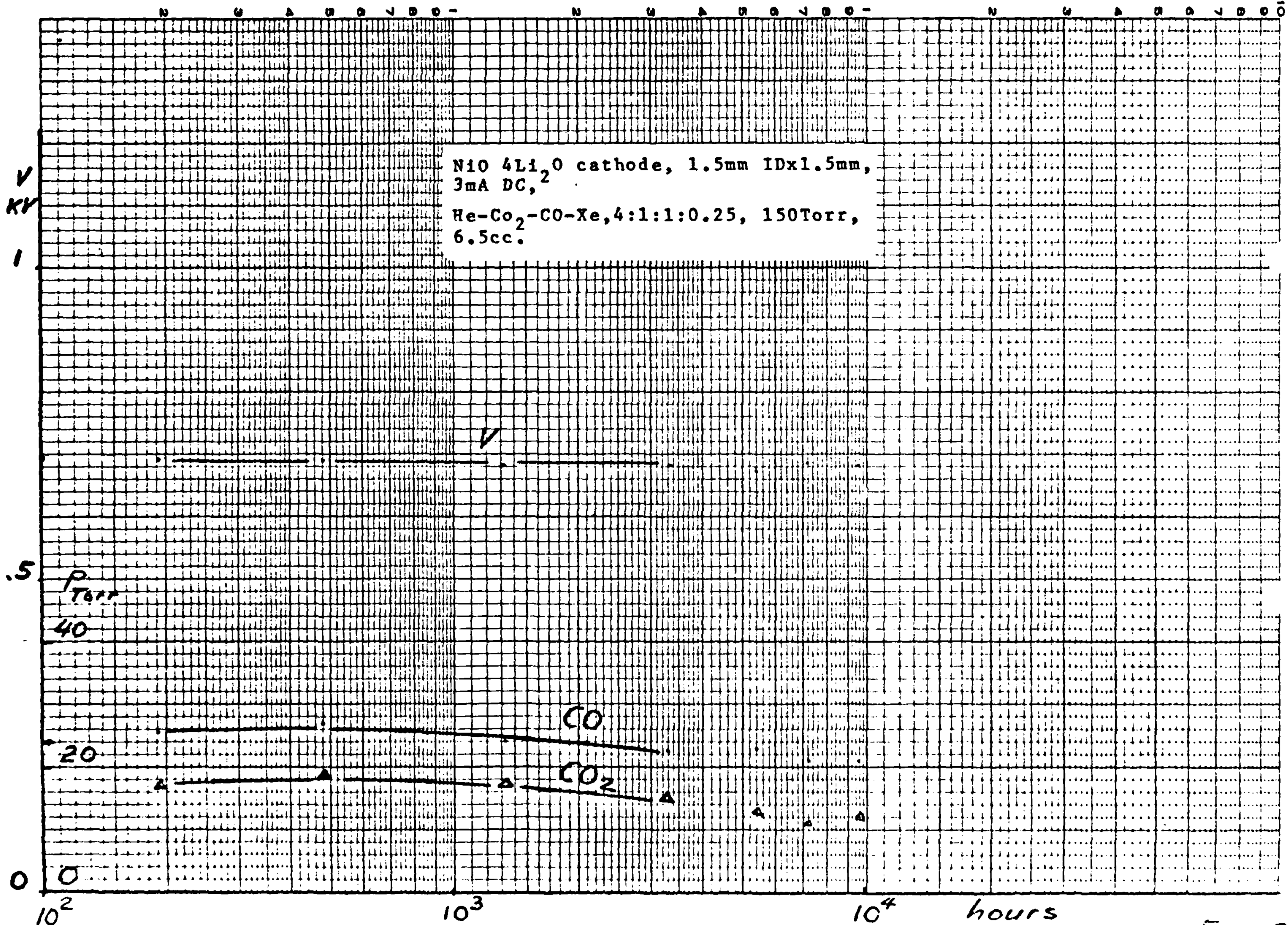
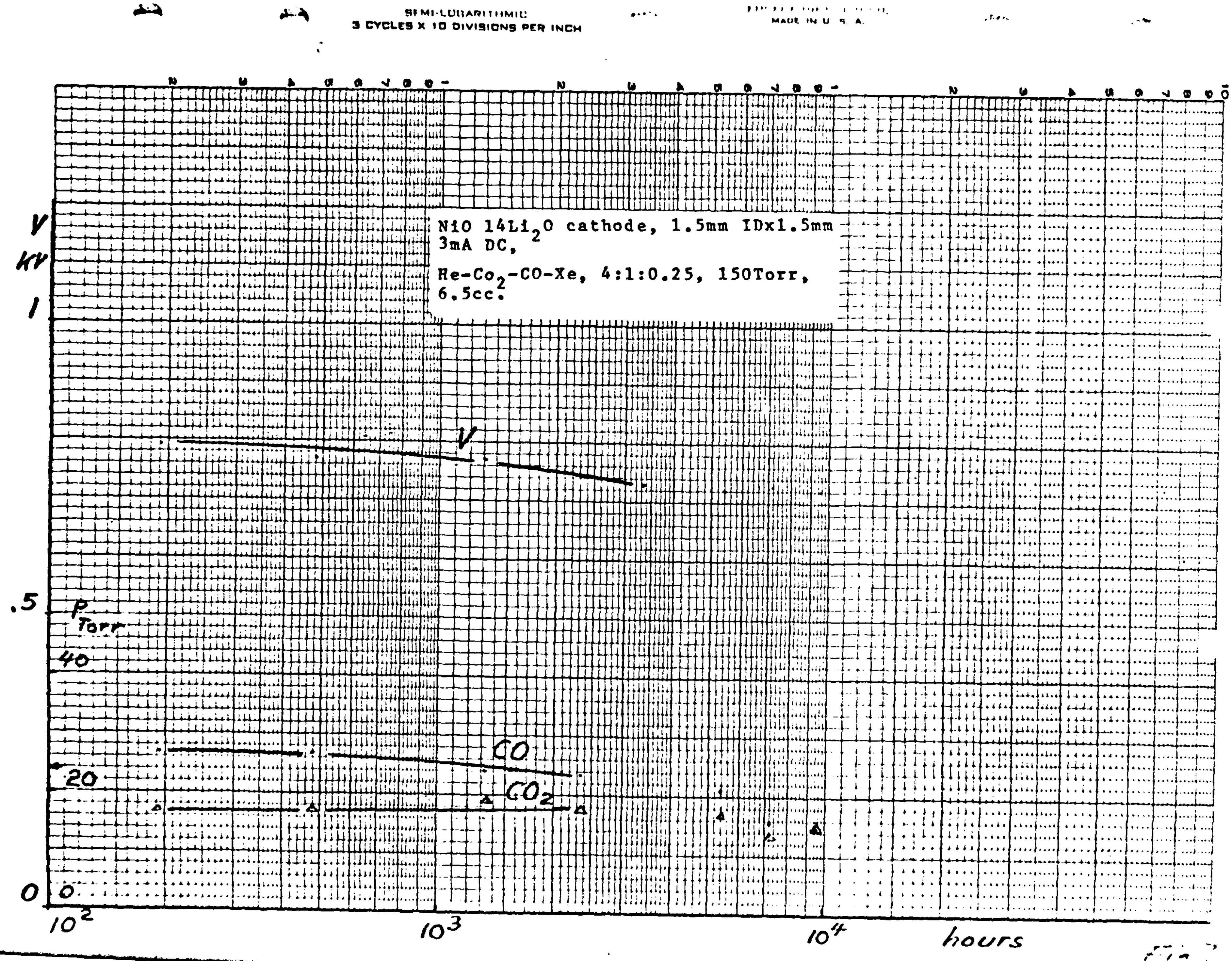
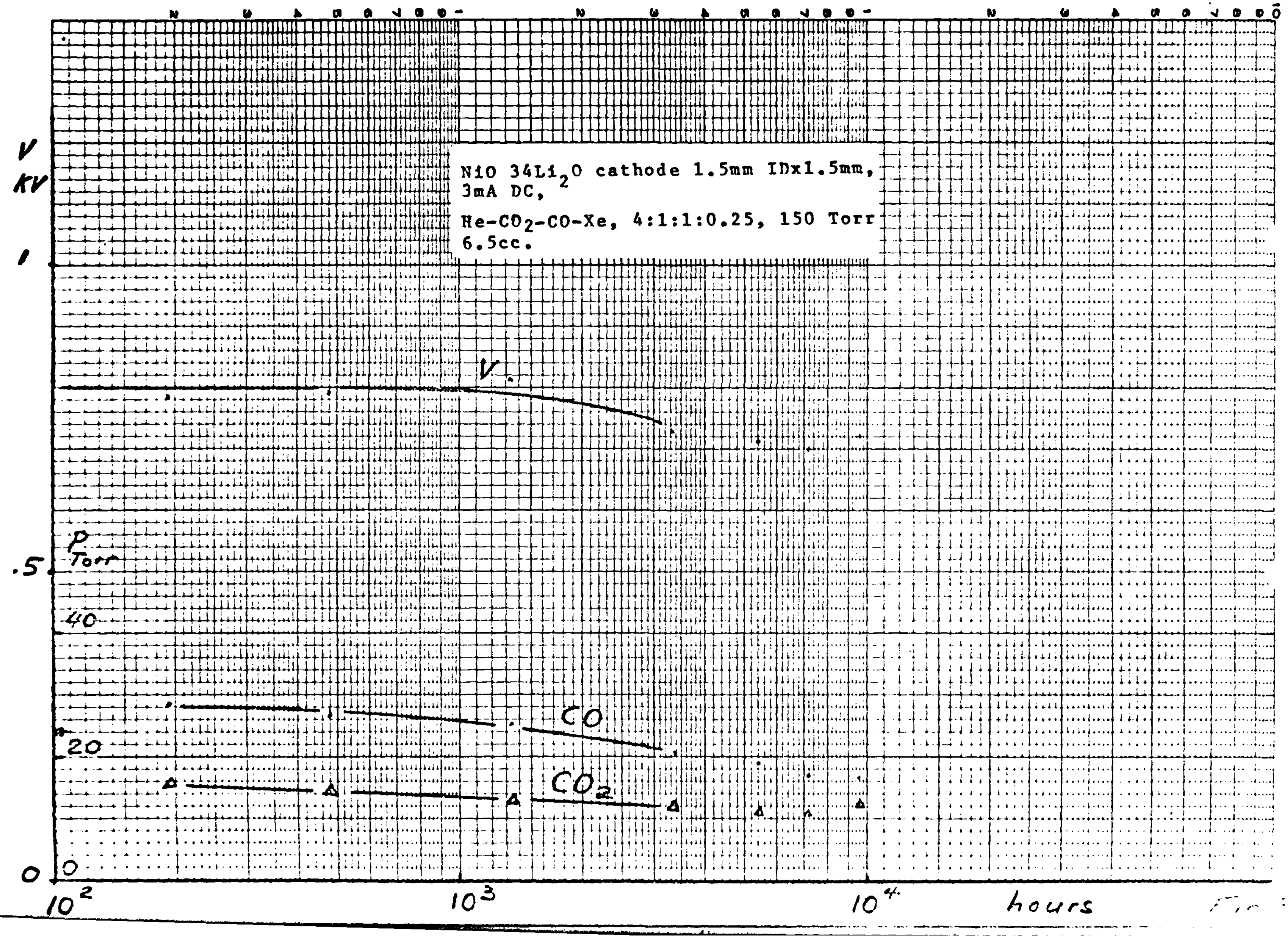


Fig. F


SEMI-LOGARITHMIC
3 CYCLES X 10 DIVISIONS PER INCH
MADE IN U. S. A.
NiO 14Li2O cathode, 1.5mm IDx1.5mm
3mA DC,
He-Co2-CO-Xe, 4:1:0.25, 150Torr,
6.5cc.
V
KV
1
.5
P Torr
40
20
0
0
V
CO
CO2
10^2
10^3
10^4
hours


V
KV
1
.5
0
P
Torr
40
20
0
NiO 34Li$_2$O cathode 1.5mm IDx1.5mm, 3mA DC,
He-CO_2-CO-Xe, 4:1:1:0.25, 150 Torr 6.5cc.
V
CO
CO_2
10^2
10^3
10^4
hours

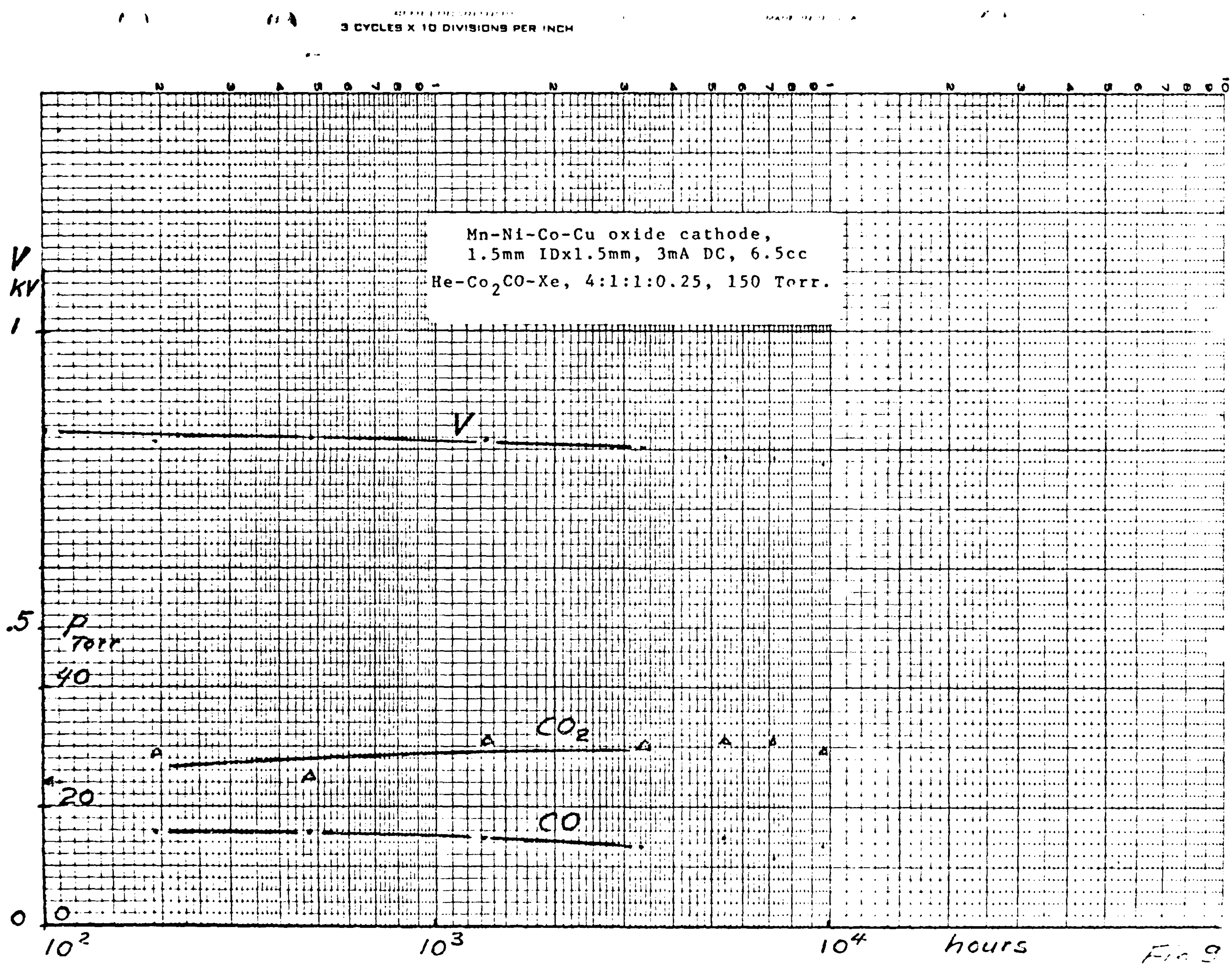

3 CYCLES X 10 DIVISIONS PER INCH
Mn-Ni-Co-Cu oxide cathode,
1.5mm IDx1.5mm, 3mA DC, 6.5cc
He-Co2CO-Xe, 4:1:1:0.25, 150 Torr.
V KV
1
.5
0
P Torr
40
20
0
V
CO2
CO
10^2
10^3
10^4
hours

Fig 9

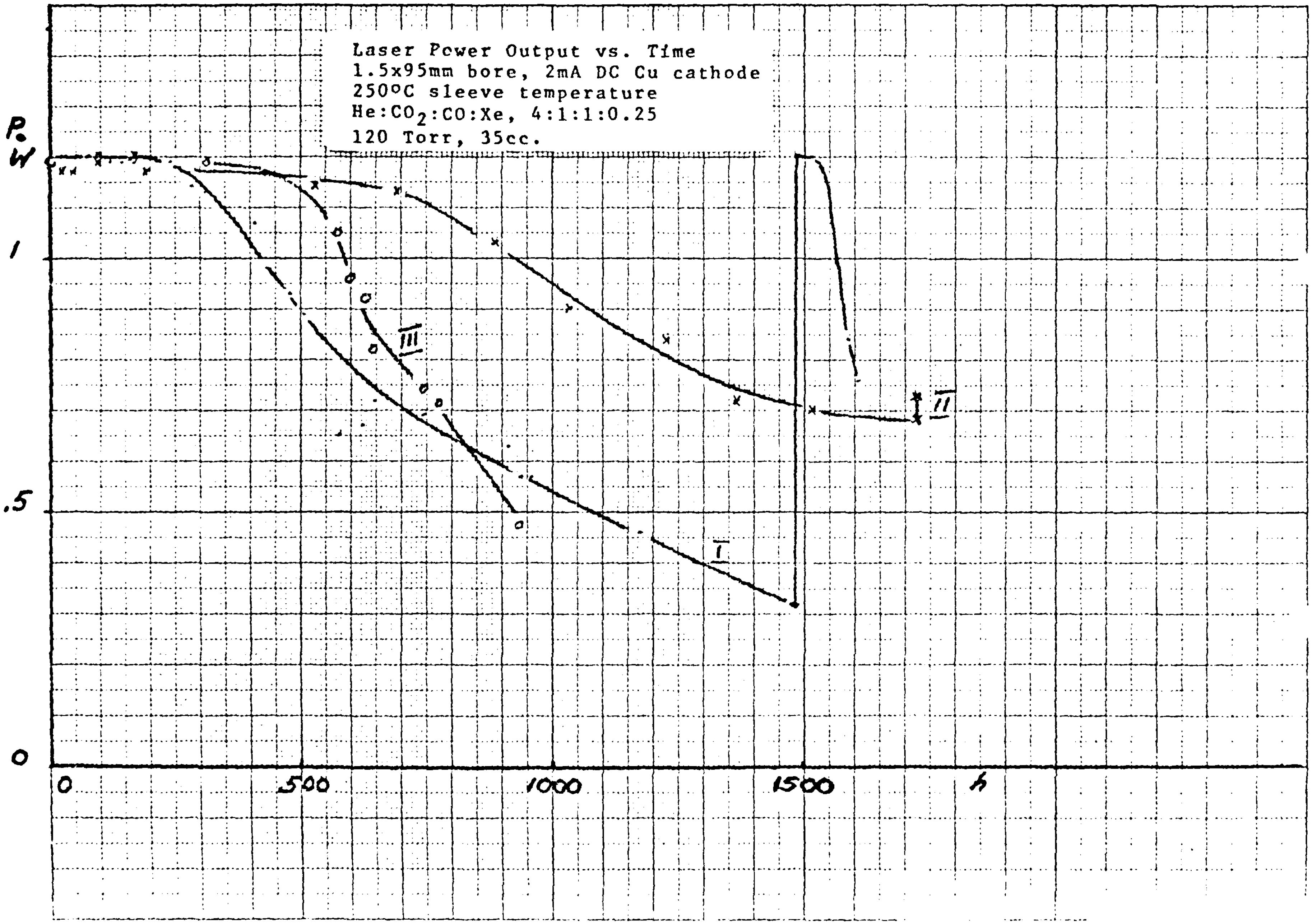

Laser Power Output vs. Time
1.5x95mm bore, 2mA DC Cu cathode
250°C sleeve temperature
He:CO_2:CO:Xe, 4:1:1:0.25
120 Torr, 35cc.
P
W
1
.5
0
0
500
1000
1500
h
I
II
III

Fig. 10

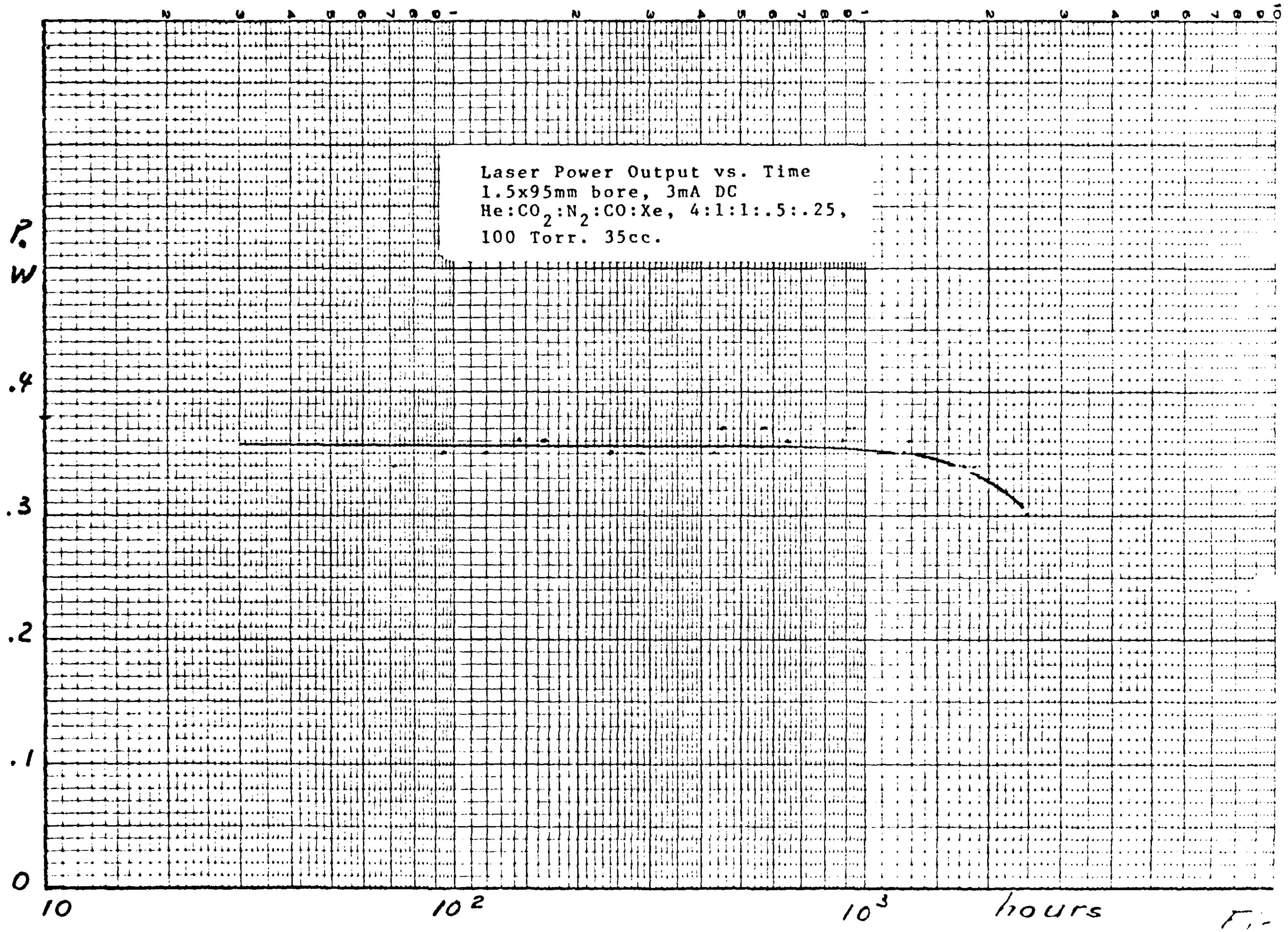
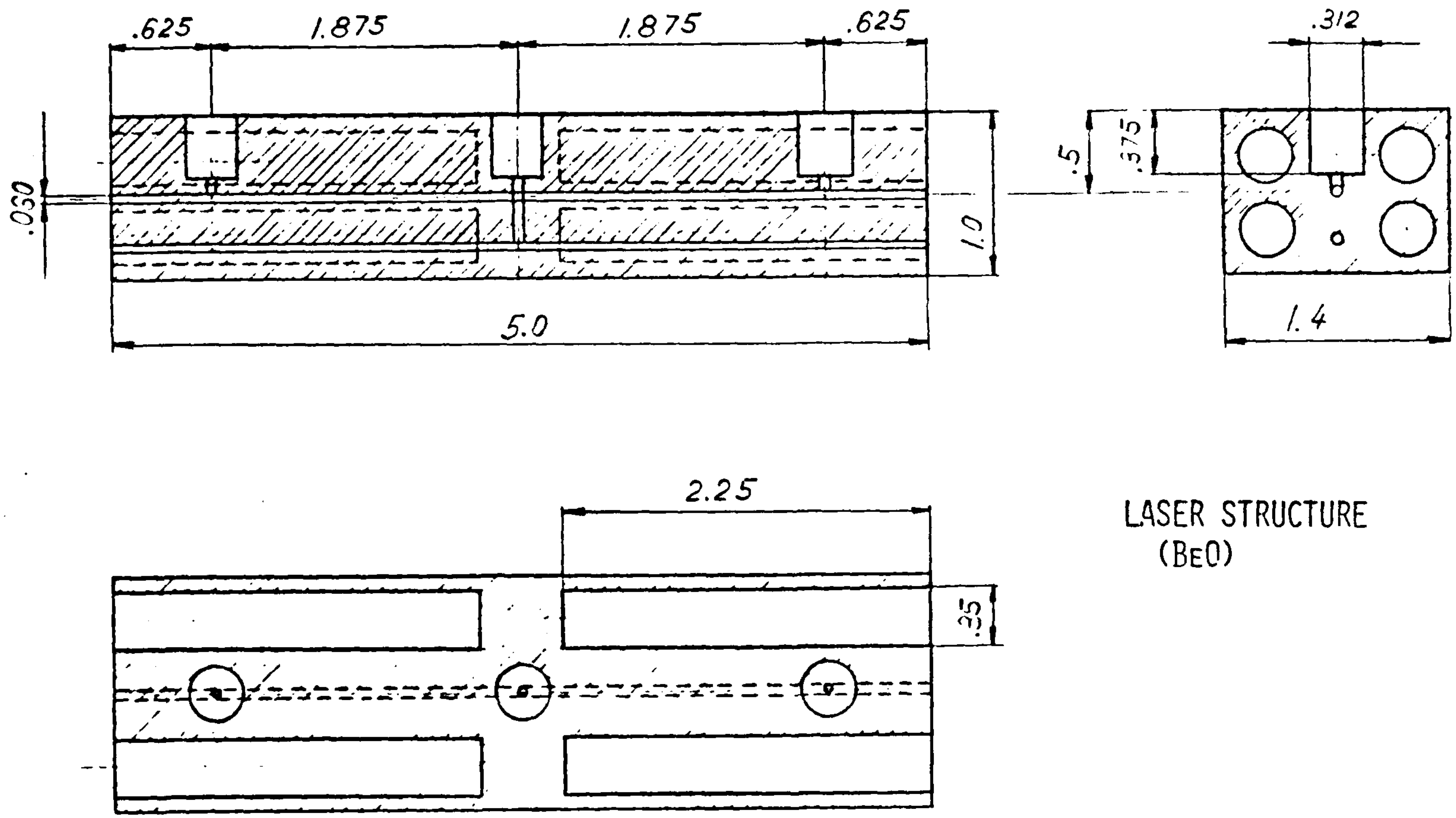

Laser Power Output vs. Time
1.5x95mm bore, 3mA DC
He:CO_2:N_2:CO:Xe, 4:1:1:.5:.25,
100 Torr. 35cc.
P. W
.4
.3
.2
.1
0
10
10^2
10^3
hours

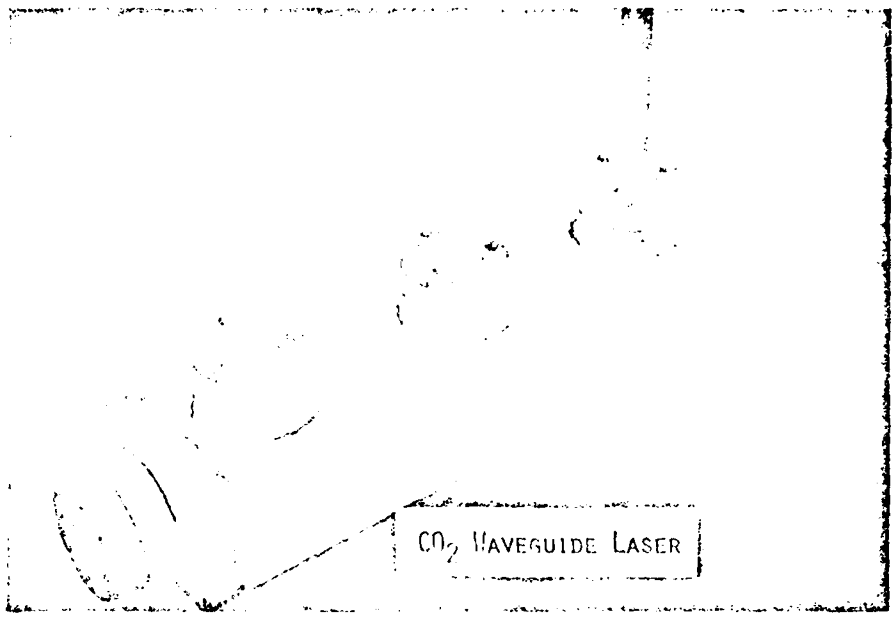
Fig. 11

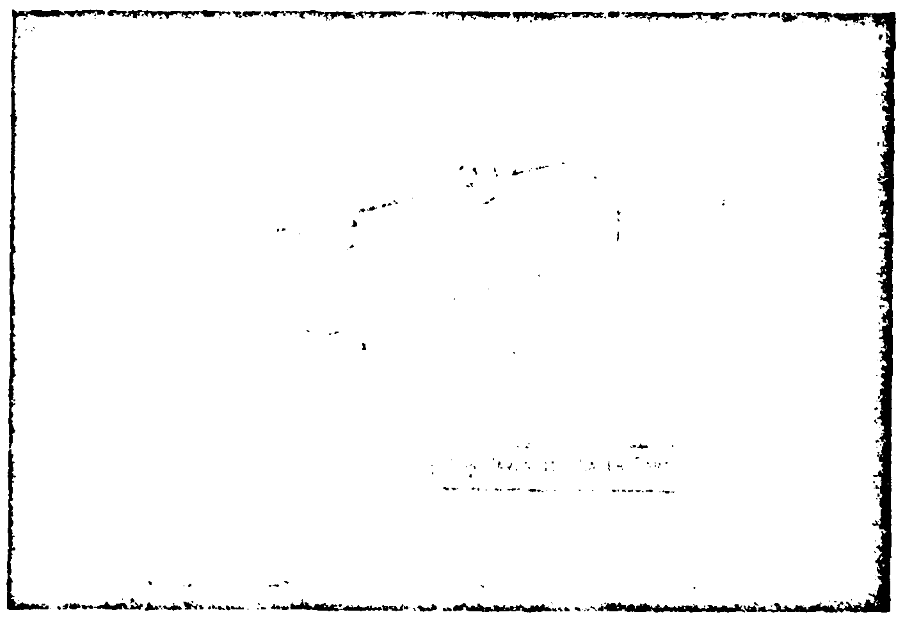


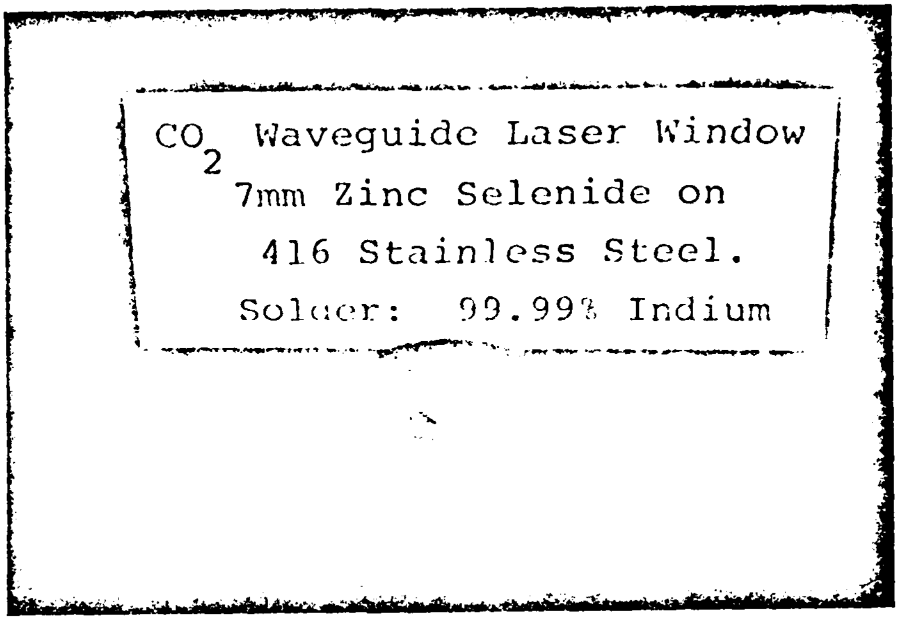
LASER STRUCTURE
(BeO)

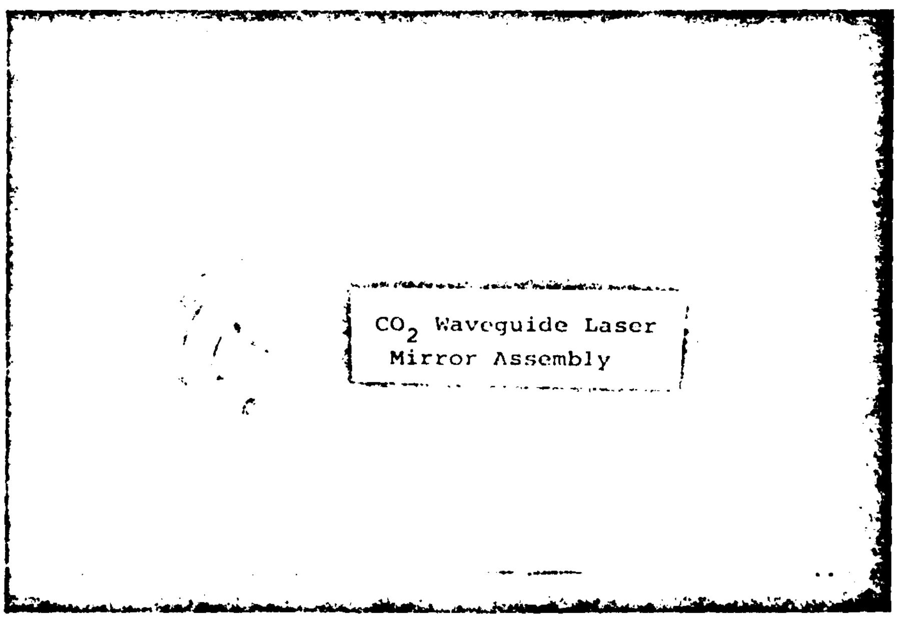
Fig. 12

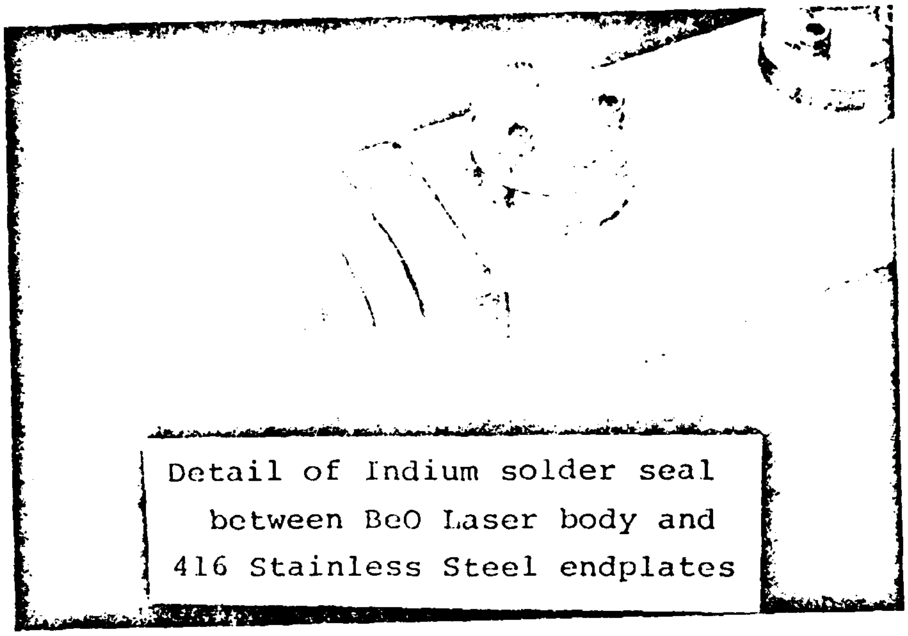
LASER STRUCTURE



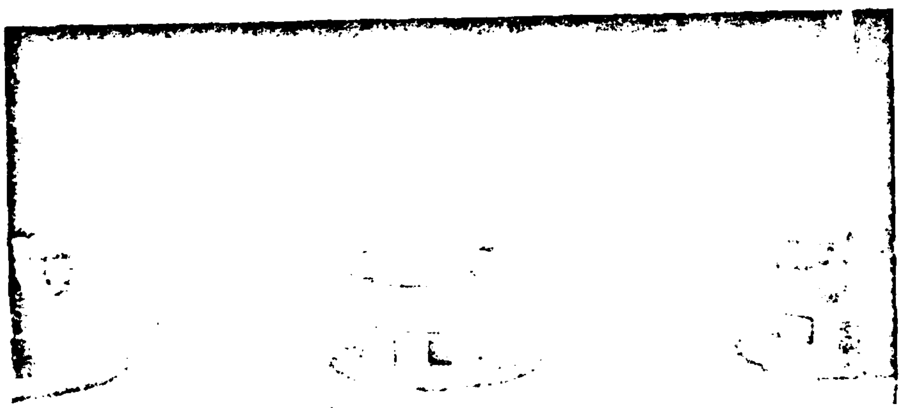



CO_2 Waveguide Laser Window
7mm Zinc Selenide on
416 Stainless Steel.
Solder: 99.99% Indium


CO_2 Waveguide Laser
Mirror Assembly



Detail of Indium solder seal
between BeO Laser body and
416 Stainless Steel endplates



Detail of Indium solder seal
between BeO Laser body and
22mm O.D. 416 Stainless Steel ring.